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Indoor and Ambient Particulate Matter Relationships during the Heating Season

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As part of the wider research topic '*Air quality over a complex topography*' the report assesses indoor pollution in a University in a residential area of Zagreb, Croatia and its relationship to ambient pollution and climatic conditions.

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1.Introduction

Particulate matter (PM), composed of solid particles and liquid droplets suspended in air, is the main constituent of polluted air in many urban regions (e.g. Ramachandran et al. 2000). It has attracted attention in recent years as it is implicated in respiratory and cardiovascular damage and, in severe cases, mortality (e.g. Görner et al. 1995). Furthermore, it has important climatic implications, affecting cloud formation and residence times, the global radiation budget and visibility (Thatcher et al. 1995; Morawska et al. 2003).

Created by a wide range of primary and secondary processes , the sources, composition and physicochemical properties of aerosols are spatially and temporally variable (e.g. Evans et al. 1999). Primary organic and inorganic particles originate from remote and urban sources including particulate matter from combustion, volcanic activity and windblown sea salt, biological material and dust. The inorganic fraction includes SO_4^{2-}, NO_3^-, NH_4^+ and Cl^- with predictable properties and behaviour (Jimenez et al. 2009). Conversely, the organic aerosol (OA) fraction contains several thousands of compounds, the majority of which remain unidentified (e.g. Evans et al. 1999).

With such diversity PM is categorised by size. Size is dependant on sources and processing and affects atmospheric behaviour and health and climatic impacts (e.g. Meng et al. 2009). Fine particles are particles up to 1µm (PM 1) and 2.5 µm (PM 2.5), generally from combustion sources. In the urban atmosphere the smallest fine fractions originate from traffic pollution and the larger from industrial processes. Coarse fractions up to 10 µm (PM 10) and larger are mechanically produced from natural and anthropogenic processes (Seinfeld et al. 2006). The finer fractions are the most damaging to health as they are easily incorporated deep into the respiratory system (Görner et al. 1995). Whilst those 1-10µm affect the upper respiratory tract (Görner et al. 1995). As it is more damaging PM 1 is the primary focus in this report, evaluating the levels of [PM 1] in a residential area of Zagreb.

As people in the developed world spend 60-90% of their time indoors (Tippayawong et al. 2009), indoor [PM 1] is important in determining personal exposure. Indoor PM sources include smoking, cooking, heating by combustion and resuspension of deposited PM by general activity within a building (e.g. Jones et al. 2000). Yet, outdoor [PM] is an important determinant of indoor [PM] (Chen et al. 2011). It enters buildings through open windows and doors and through natural and mechanical ventilation systems (Chen et al. 2011). When doors and windows are closed and ventilation systems are not used, the amount and rate of infiltration varies with building design and ambient conditions (Chen et al. 2011). Infiltration depends the magnitude of outdoor sources, PM size and characteristics, building design, air exchange rates and ambient and indoor conditions (Chen et al. 2011). It is enhanced by cracks in the building envelope, less common in modern buildings. Ambient PM 1 penetrates the building envelope more efficiently than larger size fractions because infiltration increases as particle diameter decreases (Brunekreef et al. 2005; Massey et al. 2012). Thus to understand patterns in indoor PM it is important to understand outdoor PM.

Numerous studies have investigated trends in outdoor particulate concentrations [PM] (e.g. Ramachandran et al. 2000; Hussein et al. 2006; Berghmans et al. 2009; Klaić 2012). As well as

changes in sources, ambient meteorological conditions are an important control on PM (e.g. Google maps). In Zagreb roof level [PM 1] was dependant on horizontal and vertical wind speeds, air pressure and relative humidity in Klaić (2012); (Klaić et al. 2012) and on global radiation and temperature in Klaić et al. (2012). Aside from wind speeds, these relationships were less clear at breathing height in (Klaić et al. 2012), due to close proximity to the road and a relatively short data set, thus further investigation is required to clarify climatic influences on outdoor [PM] in this region.

There have been many recent investigations into indoor air quality (e.g. Braniš et al. 2002; Brunekreef et al. 2005; Fromme et al. 2007; Stranger et al. 2009; Kearney et al. 2011; Polednik 2013; Sangiorgi et al. 2013). But to date few have investigated this in Southern Europe (Viana et al. 2011) and non in Croatia. Moreover, ambient conditions indirectly effect indoor [PM] (e.g. Braniš et al. 2005; Chen et al. 2011; Hänninen et al. 2011). But so far there has been little agreement on these relationships between studies. Thus, further investigation is required to better understand these relationships.

As the first research into indoor [PM] undertaken in Croatia this report investigates spatial and temporal variability in indoor [PM] in a residential area in Zagreb, Croatia. In addition it addresses its relationship to ambient [PM] and meteorological conditions with 1 minute mean measurement resolution.

2. Measuring site and measurements

Meteorological variables and indoor PM mass concentrations [PM] were measured at the Department of Geophysics (GFZ), Faculty of Science, University of Zagreb in a residential area of northern Zagreb (Bubble A in Fig. 1a), approximately 1.5km north-northeast from the city centre (*Donji Grad* in Fig. 1a) and 8-9km northwest of Zagreb's industrial zone (*Peščenica Žitnjak* in Fig. 1a). Outdoor [PM 1] was measured approximately 1km north-northwest of GFZ (Bubble A in Fig. 1a). Both sites are witing a moderately hilly region on the southern slope of Mount Medvednica, approximately 180m above sea level. The mountain is approximately 1 km high, spanning west-southwest and east-northeast.

Fig. 1: Maps of the measurement sites a) Satellite view of Zagreb (Liu et al. 2004). The position of the measurement sites are donated by bubble A. The city centre (Donji Grad) is approximately 1.5 km SSW of the measurement sites and the industrial estate (Pešćenica Žitnjak) is approximately 8-9 km SE of the measurement sites. b) Meteorological measurement site (Bubble B).

2.1. Meteorological data

Meteorological variables (air pressure (ρ) , air temperature (T), horizontal wind direction, horizontal (V_h) and vertical wind speed (V_w), global radiation (G) and relative humidity(RH)) were measured at GFZ using automatic meteorological station MeTA 2000 (AMeS, Brezovica, Slovenia). G and wind sensors are on the roof terrace, on the western side of the GFZ building (bubble B in Fig. 1b). This three storey building is approximately 50m long. G, horizontal and vertical wind sensors are 4.3 m, 1.8 m and 4.5 m above ground, respectively. T and RH sensors are 2m above ground in a standard meteorological station, in a grassy area north of the GFZ. The ρ sensor is inside the building, 7.5m

above ground (187.55 above mean sea level). Meteorological variables are measured at 1 second intervals and 1 minute averages are stored. North and northwest of the meteorological measurement sites are several east facing 3-5 storey, 50-70m long university campus buildings. Additionally there are smaller 1-3 storey houses and villas with sloped roofs in the area. Between the buildings are irregularly arranged trees of approximately 10 m and grassy areas.

2.2. Outdoor PM 1 mass concentration measurements

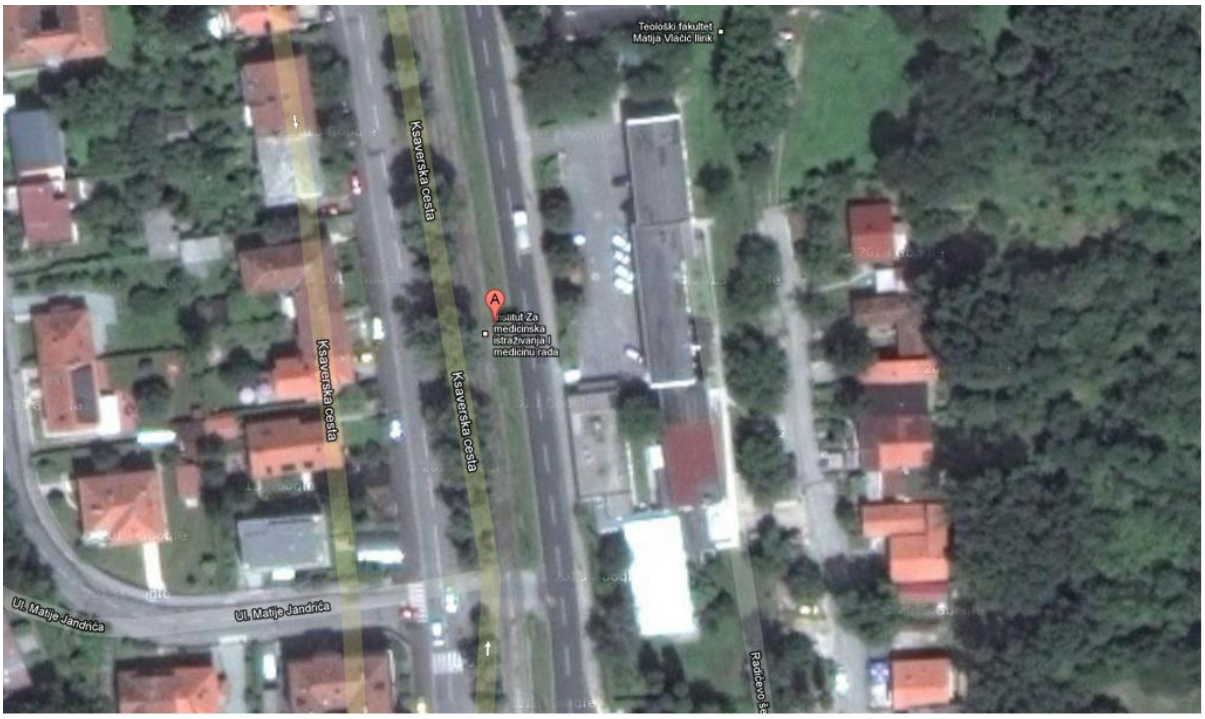

Fig. 2: Map of the ambient PM mass concentration measurement site.

Outdoor measurements of PM 1 mass concentration [PM 1] were performed at the Institute for Medical Research and Occupational Health, an urban background measuring site. The measurement station is placed approximately 50m away from the North leading side of *Ksaverska* road which has moderate traffic levels and is separated from the south leading side by trees. The surrounding area does not contain any dominant PM sources or obstacles and the ground is asphalt and grass. The closest building is 5 m high and 30 m away from the site. Measurement was performed with an LVS3 sampler (Sven Leckel, Berlin, Germany) with a Whatman^{TM} QMA quartz filter (GE Healthcare Companies,). The LVS3 is an inertial impaction with a flow rate of 2.3m^3 hr^{-1} with the inlet diameter is set for measuring [PM 1] over 24 hours. The inlet was set at 1.8m (According to EC 1999). Filters are weighed twice before, and twice after sampling (According to CSN/EN/14907 2005). A Mettler Toledo MX5 was used for weighing after 48 hours conditioning at 20°C (± 1°C) and RH 50 % (± 5 %) and again after 24 hours under the same conditions. 1 hour PM 1 averages are stored.

2.3 Indoor PM1, 2.5, 4 and TSP mass concentration measurements.

Ground floor [PM 1] was measured with the DUSTTRAK™ Aerosol Monitor, Model 8520 (TSI, Inc., Shoreview, MN, USA); first floor PM1, 2.5, 4 and TSP mass concentration measurements were measured with DUSTTRAK™ Aerosol Monitor, Model 8533 (TSI, Inc., Shoreview, MN, USA). Both monitors were calibrated at the factory. To be directly comparable to meteorological measurements the monitors was set to store 1 minute means from 1 second measurements.

Fig. 3: Location of DUSTTRAK™ monitors a) ground floor, b)first floor. Both monitors were suspended so the inlet was at average breathing height of 1.7m.

Both DUSTTRAK™ monitors were suspended 1.67m above the floor, so the inlet corresponded to the average breathing height of 1.7m (According to EC 1999), on level shelves in the corridors of the ground (fig. 3a) and first floors (fig. 3b) adjacent to the stair case. The staircase is approximately 1 m wide and 4m high and the entrance hall and landing are approximately 5 x 14 m, both have polished stone flooring. Additionally, the first floor has an approximately 1 m wide 3 m high spiral staircase connecting to the second floor. The north facing wall of both floors contains several approximately 2 x 3 m, single glazed windows, 7 on the first floor and 6 on the ground floor. There is an approximately 1 m x 0.5 m radiator the base of each window. In addition, the entrance is on the ground floor, are 2 sets of double glazed double doors, approximately 2 x 2 m.

The average number of people using the two floors on weekdays is 65 per day including 35 staff and 30 students, with arrival and leaving times varying. The busiest periods of movement are 07:00-09:00 LST with the arrival of most of the staff, 12:00-14:00 LST for lunches and 15:00-17:00 LST for people leaving the building. However, after 17:00 LST there are around 10 people in the building, leaving at

varying times up to 21:00 LST when the building is empty. Student movement within the building varies depending on lecture times and use of facilities; all students have left the building by 16:30 LST. Additionally staff from the upper level of the building (around 10 additional people) may pass through the building at any time. The floors are cleaned once daily, in the morning for the ground floor and afternoon for the first floor. Smoking is not allowed in the building but smokers congregate outside the entrance to the building.

Activity is very low during the weekend. Officially there is one member of staff in the building 07:00-14:00 LST on Saturdays and no one on Sundays. However, occasionally staff members may come to the building at any time over the weekend and this is not recorded. Central heating for the building is set to 21°C 06:00-21:00 and 17°C 21:00-06:00 on weekdays and 17°C on weekends. For Christmas holiday period the temperature was 19°C for 21:00 LST 22.12.2012. - 06:00 LST 02.01.2013. As measurement were taken during the winter time, windows closed in the immediate vicinity of the DUSTTRAK™ monitors, yet during the period windows of the offices on the adjoining corridor were occasionally opened for air exchange or temperature regulation.

The closest roads to building are *Horvatovac* and *Bijenička*, 50 and 100 m away respectively. Traffic levels were not investigated in the present study, but *Horvatovac* tends to how low intensity traffic whilst (Klaić et al. 2012) details the average traffic behaviour of *Bijenička* road as low speed, low-moderate intensity, whilst traffic levels are low during non working hours .

2.4 Data

For meteorological and indoor [PM] data 75 days of winter measurements from 18.11.2012 – 30.01.2013 were obtained. Outdoor data is for 70 days the period 18.11.2012 – 25.01.2013. For the majority of analysis Indoor data was separated into 3 categories depending on the level of activity in the building, 1. weekdays, 2.Saturdays, 3. Sundays and Christmas holidays 22.12.2012-02.01.2013. These categories of data were analysed separately to activity within the building effects on temporal variation. Outdoor [PM] data was categorised similarly due to the variability of traffic between 1.weekdays, 2. Saturdays, 3. Sundays, the Christmas holiday period was not separated as working days in non academic proffessions were not affected. Due to construction works in the building 3.12.2012-13.12.2012 indoor [PM] levels were considerably higher than for any other time in the measurement period between 08:00 LST on 3.12.2012 to 08:00 LST 17.12.2012 so this period was excluded from the analysis of indoor data.

3. Results and Discussion

3.1 PM 1 mass concentrations [PM 1]

Descriptive statistics for [PM] are in table 1. The mean ambient [PM 1] value is 0.0339 ± 0.233 mg m^{-3}. Due to regional and seasonal differences in PM sources, macro and micro meteorology and different experimental design it is difficult to make quantitative comparisons with the literature. However outdoor [PM 1] is comparable with the average of 10 winter road measurements (0.0387 mg m^{-3}) on *Bijenička* road in Klaić et al. (2012); estimates in Mol, Belgium for cyclist (0.0374 mg m^{-3}) exposure based on 7 experiments (Berghmans et al. 2009) and winter weekday daytime averages of ambient [PM 1] at a state air-quality monitoring network station in Prague, Czech Republic (0.039 mg m^{-3}) (Braniš et al. 2005).

Mean [PM 1] indoors (0.0579 - 0.0633 mg m^{-3} (table 1)) was within the range of winter values in occupied classrooms in Lublin, Poland (0.047 – 0.118 mg m^{-3}) averaged from 1 day of continuous measurements (Polednik 2013). Values are also similar to average non-winter weekday measurements (0.0656 mg m^{-3}) in classrooms in Delhi, India (Goyal et al. 2009). All of the above studies were similar to the present as there were no significant indoor sources. Values for the other size fractions are similar to [PM 1] and are discussed in section 3.5.

Conversely, mean indoor [PM 1] (0.0579 – 0.0633 mg m^{-3}) for the same period was considerably higher than for weekdays in (Braniš et al. 2005) (0.0137 mg m^{-3}). Differences may be partly due to the difference in monitor height as in (Braniš et al. 2005) the inlet was set to sitting breathing height. Along with the factors discussed for outdoor [PM 1] varying building types and activities indoors also make comparisons different. Additionally, gravimetric aerosol monitoring equipment was used in (Braniš et al. 2005), which has been reported to give lower estimates of [PM 1] than photometric methods used in this study.

Table 1: Descriptive statistics [PM] (mg m^{-3}) for the period 18.11.2012-30.01.2013 for outdoor (out), ground floor (GF) and first floor (FF) measurements and the in – out ratio (I/O) based on GF and outdoor measurements.

	mean	max	min	median	mode	st. dev	variance
[PM 1] out	0.0339	0.127	0.003	0.028	0.018	0.0233	0.0005
[PM 1] GF	0.0579	0.314	0	0.043	0.011	0.0515	0.0027
[PM 1] FF	0.0633	0.303	0	0.050	0.016	0.0509	0.0026
[PM 2.5] FF	0.0634	0.303	0	0.050	0.016	0.0509	0.0026
[PM 4] FF	0.0636	0.303	0	0.050	0.016	0.0509	0.0026
[PM 10] FF	0.0641	0.303	0	0.050	0.016	0.0511	0.0026
[TSP] FF	0.0659	0.427	0	0.052	0.016	0.0515	0.0026
I/O ratio	1.64	9.46	0	1.48	0	0.905	0.819

3.2 Atmospheric conditions and impacts on ambient and indoor [PM]

3.2.1 Meteorological conditions

Table 2 shows descriptive statistics atmospheric conditions for the measurement period (18.11.2012 – 30.1.2013) and Fig. 4 shows average diurnal variation for the same period. All of the variables follow the expected diurnal trend in agreement with (Klaić et al. 2012).

Table 2: Descriptive statistics for meteorological variables for the period 18.11.2012-30.01.2013 for temperature (T), air pressure (P), relative humidity (RH), global radiation (G), vertical wind velocity (V_w) and horizontal wind velocity (V_h).

	mean	max	min	median	mode	st. dev	variance
T (◦C)	3.81	16.30	-5.50	3.00	0.10	4.41	19.44
P (hPa)	993.40	1012.90	970.90	994.60	995.10	8.86	78.56
RH (%)	89.11	100.00	49.00	94.00	100.00	12.03	144.66
G (J cm^{-1})	30.47	553.00	0.00	0.00	0.00	76.06	5785.46
V_w (cm s^{-1})	9.58	181.00	-63.60	3.90	0.60	17.68	312.46
V_h (m s^{-1})	0.98	15.50	0.00	0.50	0.10	1.28	1.64

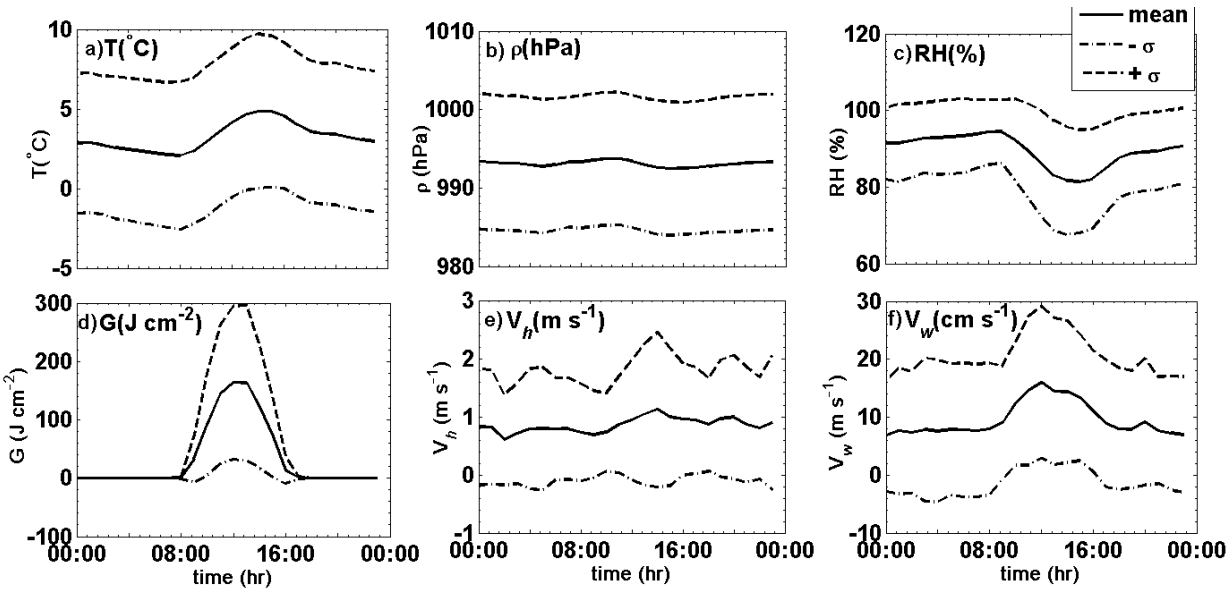

Fig. 4: Mean diurnal variation (solid line) +/- standard deviation (dashed and dot-dash line respectively) in meteorological parameters for the measurement period 18.11.2012 – 30.01.2013. a) Temperature, T (°C); b) air pressure, ρ (hPa); c) relative humidity, RH (%); d) global radiation, G (J cm^{-2}); e)Horizontal wind speed, V_h (m s^{-1}); f) vertical wind speed, V_w (cm s^{-1}). Note the x scale is the same for all subplots.

3.2.2 Atmospheric conditions impact on ambient [PM 1]

Fig.5. shows the relationship between minute mean meteorological conditions on minute mean ambient [PM 1]. With lower temperatures (-14 - -2°C) [PM 1] is higher (Fig. 5a.), probably due to more particulate emissions from the central heating plant in the west and south – east of the city. Furthermore, at colder temperatures there is more atmospheric stability. Consequently, reduced turbulence and lower boundary layer height increase the residence time of pollutants and allow secondary species to form (e.g. Braniš et al. 2009). Furthermore, pollutants partition into the particle phase at lower temperatures. Higher [PM 1] at colder temperatures agrees with a number of studies reporting higher ambient [PM 1] in winter (e.g. Evans et al. 1999; Vecchi et al. 2004; Hussein et al. 2006; Polednik 2013), but the relationship with temperature may depend on the temperature range in question. It is expected that the relationship with temperature in summer would be positive as summer conditions facilitate photochemical production of PM.

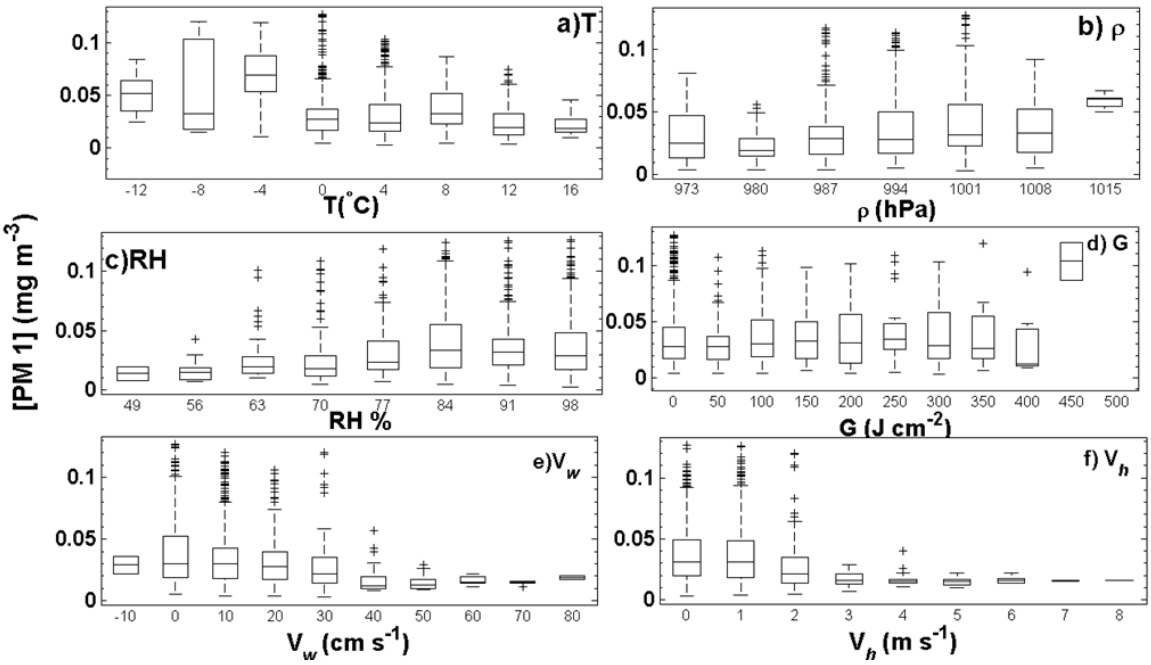

Fig. 5: Boxplots of ambient PM 1 mass concentrations ([PM 1] (mg m^{-3})) and meteorological parameters for the measurement period 18.11.2012 – 25.01.2013. a) Temperature, T (°C); b) air pressure, ρ (hPa); c) relative humidity, RH (%); d) global radiation, G (J cm^{-2}); e) vertical wind speed, V_w (cm s^{-1}); f) Horizontal wind speed, V_h (m s^{-1}). Note the x axis for all subplots are the same scale.

Additionally, the T relationship in this study is not linear. This may be explained by the rarity of higher temperatures during the winter period providing less data for higher temperatures or perhaps by the interaction of some other climatic or source variables. However, summer results from roof measurements at GFZ in (Klaić et al. 2012) also showed a wave-like pattern with a increase in [PM 1] below 4.5°C and again around 12°C and 30°C. So the temperature affect may be non linear. But inter-study differences in aerosol monitor height and seasonal differences should be taken into account. Firm conclusions cannot be drawn from this without cross seasonal measurements under the same conditions.

Higher [PM 1] coincide with higher ρ (> 1000 hPa) (Fig. 5b.) in agreement with roof measurements in Klaić et al. (2012). As with temperature, this increase may be associated with stable atmospheric stratification which may lead to temperature inversions close to the ground, increasing residence time of pollutants, facilitating particle formation. At lower ρ the relationship is less clear, in a wave shape. This is comparable with (Hussein et al. 2006) which did not find a relationship between [PM 2.5] and ρ. However, Hussein et al. (2006) varies from the present study in measuring the number concentration rather than mass concentration of a different size fraction, measurement height and temporal scale so direct comparison is not possible.

Again, for RH (Fig. 5c.) there is a wave-like pattern, Despite this pattern the general trend is an increase in [PM 1] with RH similar to Klaić (2012); Klaić et al. (2012), which is expected due to greater gas to particle partitioning of volatile aerosols with increasing RH. The highest [PM 1] occur around 85% RH, after which the slight decrease may be explained by particle removal via precipitation or fog as in (Klaić 2012; Klaić et al. 2012). Similarly slight (-10%) reductions in [PM 1] during precipitation were reported in (Vecchi et al. 2004). Again this is in contrast to (Hussein et al. 2006) who found no relationship with RH.

There is no clear relationship between G and [PM 1] (Fig. 5d.), similarly to road measurements in (Klaić et al. 2012). However, as measurements in the present study and in road measurements (Klaić et al. 2012) were performed during short, often cloudy days in winter the range of G values is relatively small (30 – 533 J m^{-2} in the present study) so the suggested cross seasonal study would give a wider range of values and a better understanding of this parameter.

The wave patterns apparent between T, RH and ρ and [PM 1] and the apparent lack of relationship between G and [PM 1] may be explained by variation in pollutant sources resulting from changes in wind speed and direction and long and short distant transport of pollutants as discussed below. For example in the same region for SW, W, WNW and WSW winds there was not a concurrent decrease in [PM 1] with the highest RH (Klaić et al. 2012). The same study found the relationship between G and [PM 1] altered depending on wind directions. Additionally, the interrelationship between the different meteorological variables and PM source variability may make relationships of individual climatic variables less clear. Such analysis is beyond the scope of this study but comparing source fluctuation, meteorological variation and [PM 1] could be used in future investigation.

For vertical wind (Fig. 5e.) the highest [PM 1] are associated with negative to low wind speeds (-10 - 5 cm s^{-1}), during which stable conditions may facilitate pollutant trapping, whereas concentrations decrease above this. Thus there is dilution of pollutants with increased convection leading to efficient transport of pollutants to greater heights as with road measurements in Klaić et al. (2012). However, unlike Klaić et al. (2012) the present study does not show a decrease in [PM 1] with decreased subsidence, likely due to aerosol monitor being further from car exhaust pollutant sources in the present study.

There is a similar pattern for horizontal wind speeds (Fig. 5f.), with lower [PM 1] for faster winds (> 2 m s^{-1}). This suggests that advection of local pollutants is dominant at lower speeds, whilst at higher wind speeds the concentrations are lowest due to dispersion similar to Klaić et al. (2012). Similarly,

substantial decreases in [PM 1] (20-30%) were related to wind speeds > 2 m s^{-1} in Milan (Vecchi et al. 2004). Again, different relationships with horizontal wind speed and [PM 1] are expected for the differing wind directions discussed below, but this detail of analysis is beyond the scope of this study.

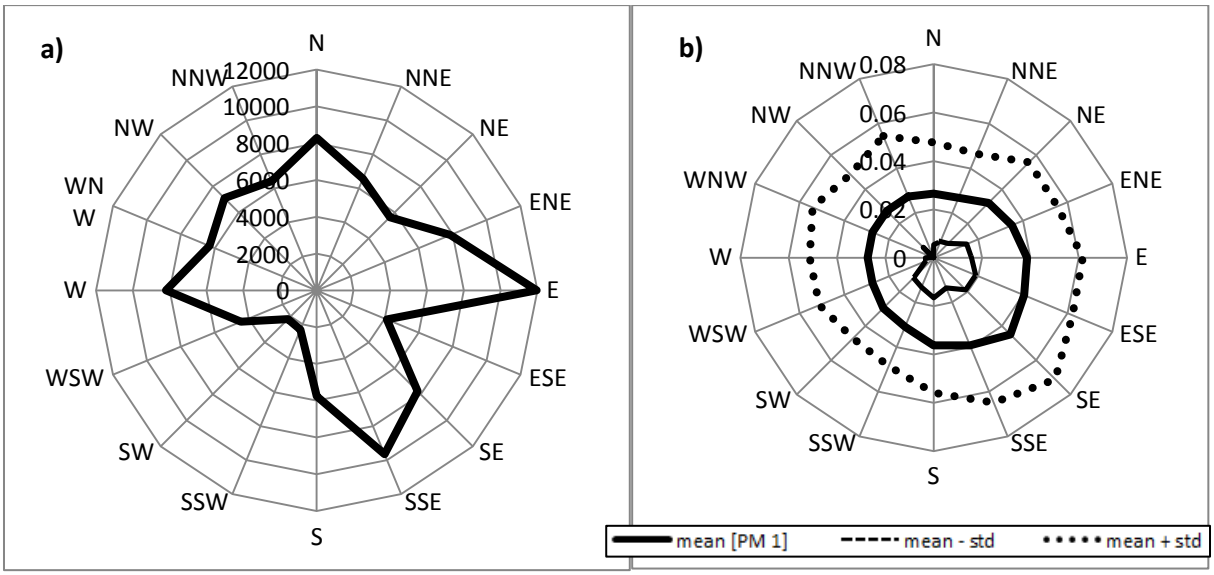

Fig. 6: Horizontal wind directions at 22.5° intervals for the measurement period 18.11.2012 - 31.01.2013 and relationship with PM mass concentrations a) absolute frequency of wind directions b) PM mass concentration ([PM 1] (mg m^{-3})) variation with wind direction.

Horizontal wind direction is important (Fig. 6.) for transporting local and long distance pollution Fig. 6a shows the absolute frequency of wind directions over the measurement period, whilst Fig. 6b shows the hourly mean [PM 1] with the hourly wind direction at 22.5° intervals. Long distance and regional transport of pollutants and cleaner air is controlled by the wind direction. In agreement with (Klaić et al. 2012), the lowest [PM 1] are associated with N winds (0.0265 mg m^{-3}), when cleaner air from Mt. Medvednica is transported to the site. Furthermore, N winds are colder and drier than the other directions (Klaić et al. 2012) thus not facilitating the production of [PM 1] which is favoured in more humid, warm conditions. This is closely followed by WSW – NNW which all have a mean [PM 1] of (0.0274 mg m^{-3}), for NNW and NW winds are also associated with clean downslope winds from Mt. Medvednica (Klaić et al. 2012). Low concentrations were not observed for WSW-WNW in (Klaić et al. 2012), this difference may be explained by the measurement site in this study being more isolated from long distance transport than (Klaić et al. 2012) as the surrounding area contains the higher altitude upper town and hilly areas.

The highest [PM 1] are associated with SE winds (0.0449 mg m^{-3}), due to long distance transport of pollutants from the industrial estate in SE Zagreb, additionally SE winds are humid in Croatia (Klaić et al. 2012) increasing gas to particle partitioning. Whilst the distribution of wind directions and [PM 1] in the present study is similar to other research in this area, the SE wind component has a higher concentration than in Klaić (2012) and roof measurements (Klaić et al. 2012) with 0.0398 mg m^{-3} and 0.0350 mg m^{-3}, respectively. Differences may be explained by this measurement period being restricted to

winter, when the production of central heating is the highest whilst the other data sets are from spring (Klaić 2012) and spring and autumn (Klaić et al. 2012).

3.2.3 Atmospheric conditions impact on indoor [PM 1]

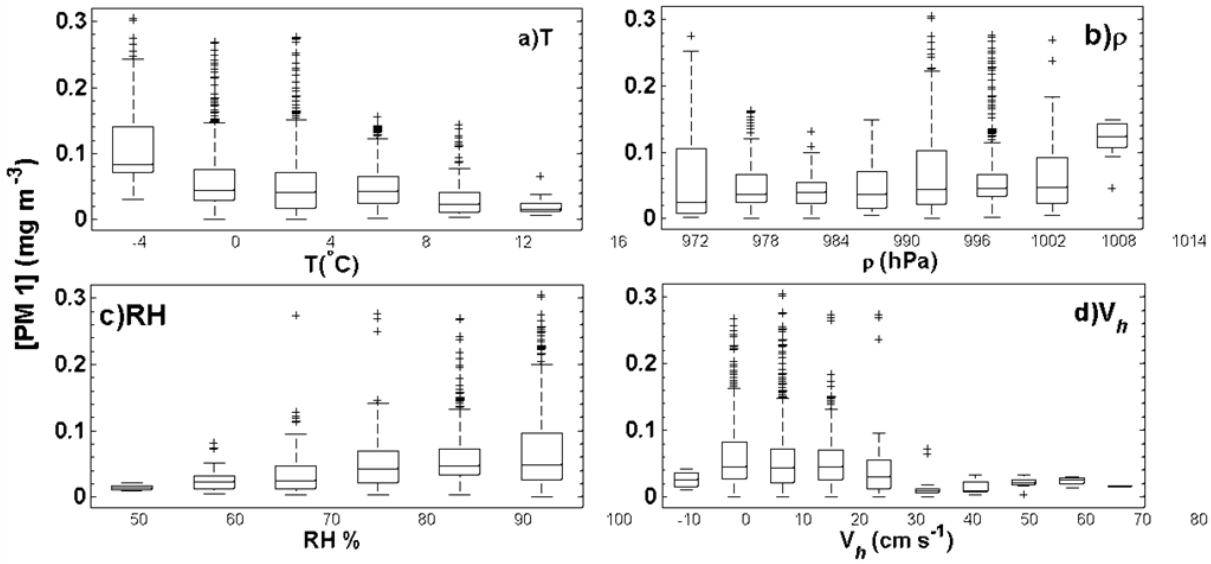

Fig.7: Boxplots of indoor PM 1 mass concentrations ([PM 1] (mg m^{-3})) and meteorological parameters for the measurement period 18.11.2012 – 30.01.2013. a) Temperature, T (°C); b) air pressure, ρ (hPa); c) relative humidity, RH (%); d) global radiation, G (J cm^{-2}); e) vertical wind speed, V_w (cm s^{-1}); f) Horizontal wind speed, V_h (m s^{-1}). Note the x axis for all subplots are the same scale.

Fig. 7. shows the indirect relationships between ambient conditions and indoor [PM 1], similarly to outdoor [PM 1] the highest concentrations are associated with the lowest temperatures. This is likely to be due to infiltration of the higher ambient concentrations associated with cold temperatures. Additionally thermal lifting of [PM 1] indoors may occur with the increased central heating required to maintain comfortable indoor temperatures. Temperature gradients between inside and outside affect the rate of infiltration of PM; when outdoor temperatures are cold the T gradient is high, thus infiltration through the building envelope may increase due to thermophoresis (Chen et al. 2011).

In agreement with the present study, an inverse relationship with temperature was reported in classrooms (Goyal et al. 2009) and homes (Massey et al. 2012) in N. Central India. The relationship was not perturbed by human activity in (Goyal et al. 2009), yet this data was for all seasons unlike the present study and indoor sources were present in (Massey et al. 2012). Furthermore, a strong negative correlation with small size fractions and temperature was only observed during daytime in schools in Chang Mai, Thailand with the relationship reversed at night time (Braniš et al. 2002). However, these were naturally ventilated buildings and climatic differences between Croatia, India and Thailand make comparison difficult.

Furthermore, this relationship with temperature and [PM 2.5], was only apparent in summer and not winter in classrooms in Munich (Fromme et al. 2007). But, the aforementioned study used gravimetric methods rather than photometry and there may be differences in infiltration between [PM 2.5] and [PM 1]. Similarly, Braniš et al. (2005) did not find a significant relationship between

temperature and indoor [PM 1] in winter measurements, this may be explained by their 12 hour measurement resolution not highlighting subtle shifts in temperature and [PM 1].

Moreover, Yao et al. (2009) found a non-linear dependence on ambient temperatures for [PM 1] in American houses with infiltration increasing with both higher and lower temperatures, greatest at 20°C. In winter, lower ambient T was also associated with high [PM 1] in a gym in Prague, whilst there was a positive correlation between the variables in summer due to increased smog formation (Braniš et al. 2009). Seasonal differences may also be impacted by other climatic variables, which are discussed below. To further investigate the effect of temperature on indoor [PM 1], seasonal variation and human activity an investigation into diurnal and seasonal variation in temperature [PM 1] relationships is required.

Like outdoor measurements, the ρ and [PM 1] relationship (Fig. 7.b), is wave-like and the highest [PM 1] is related to the highest ρ. Again this similarity may demonstrate infiltration of ambient [PM 1] indoors. Similarly to temperature, ρ gradients between the inside and outside may enhance the infiltration on ambient [PM 1], yet this is difficult to quantify due to the interaction of other variables such as wind direction and speed (Chen et al. 2011; Hänninen et al. 2011). Simultaneous measurement of indoor and outdoor pressure would clarify this.

There is a clear increase in [PM 1] with RH (Fig. 7.c), again this is similar to the ambient relationship but [PM 1] decrease at the highest RH is less apparent as precipitation events will not affect the indoor [PM 1] as strongly. (Braniš et al. 2005) also found a strong positive correlation between ambient RH and indoor [PM 1] and also with indoor RH. Further investigation of the indoor RH would help to quantify this as the RH gradient between the inside and outside will cause phase changes for volatile [PM 1]. Also, drying of particles at lower RH, will reduce particle mass. For small size fractions, there was also a positive correlation with RH during daytime in (Braniš et al. 2002). Furthermore the range of ambient RH in the present study (49 – 100%) is comparable to Braniš et al. (2002) (43-96%). It is difficult to isolate the impact of temperature and RH as they are interrelated so presumably the interaction of temperature or other variables accounts for the night time difference, again diurnal investigation would help understand this. Conversely, no significant relationship was found between RH and [PM 1] in Massey et al. (2012), but this investigates 24 hour means of [PM 1] and RH and indoor - outdoor ratios not absolute concentrations which may explain some differences

There is a clear negative correlation between horizontal wind speed and indoor [PM 1] (Fig. 7.c) explained as the indirect effect of outdoor air providing a source of [PM 1] or its precursors and the resulting ρ differences affecting the infiltration of outdoor air. This relationship was also seen in a school gymnasium in Lublin, Poland (Braniš et al. 2009) and in classrooms in Delhi, India (Goyal and Khare 2009). But whilst classrooms were occupied in the relationship was not seen. More activity than in the current study in would occur in classrooms so this may account for the difference. Additionally, there was no correlation with ρ found in Braniš et al. (2002).

The meteorological variables and indoor [PM 1] were analysed for separately days, Saturdays and Sundays and holidays to account for human activity influences on the relationships (data not shown) but no marked difference was seen. The majority of indoor data was from weekdays (47 out of 75 days) leaving small data sets for the Saturday and Sunday and holiday analysis. A more detailed

analysis separating days and evenings for a longer data set is required to further investigate the impact of activities in the building. Temporal variations in [PM 1] are discussed in section 3.3. Furthermore, to gain a clearer understanding of the causes of changes in response to ambient conditions, simultaneous analysis of indoor ρ, T, RH may be useful.

3.3. Temporal variations in [PM 1]

3.3.1 Temporal variations in ambient [PM 1]

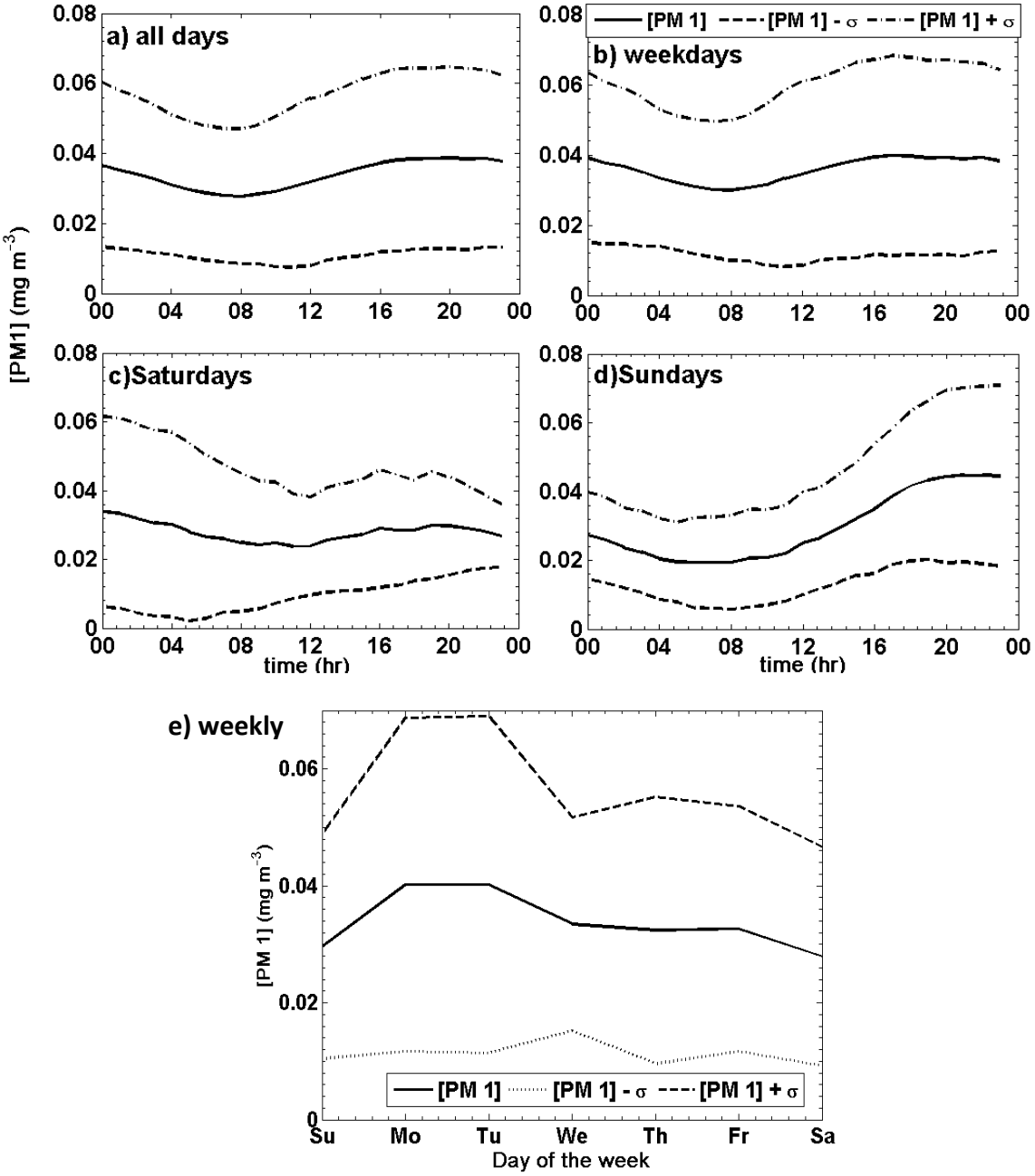

Fig. 8:Temporal variation in PM 1 mass concentrations ([PM 1] (mg m^{-3})) 18.11.2012 – 25.01.2013. a-d) diurnal variation, mean [PM 1] (solid line), mean + standard deviation (dashed line), mean – standard deviation (dash – dot line). a) All days, b) weekdays, c) Saturdays, d) Sundays. e)Weekly variation in PM 1 mass concentrations ([PM 1] (mg m^{-3})) 18.11.2012 – 25.01.2013, Mean (solid line), mean + standard deviation (dashed line), mean – standard deviation (dash – dot line).

Diurnal variations in ambient [PM 1] are shown in (Fig. 8) for the entire 70 day measurement period (Fig. 8a) and for weekdays only (Fig. 8b), the mean diurnal variation is similar, probably because the largest proportion of the measurement period was weekdays (50 of the 70 days). For weekdays peak concentration is 0.0398 ± 0.0260 mg m^{-3} at 17:00 LST and there is a decrease with minor fluctuations in concentrations to a minimum of 0.0190 ± 0.0301 mg m^{-3} at 07:00 LST, a 52% difference between the maximum and minimum. The post 07:00 LST rise is presumably associated with the morning traffic and the continued rise until around 17:00 evening is likely due to daytime traffic and photochemical production of PM with daylight and other atmospheric conditions. Similarly to road measurements in (Klaić et al. 2012) the effect of typical shallow boundary layer conditions during the night is not apparent, most likely due to the predominance of source reduction and interaction of other climatic variables. The range of diurnal mean concentrations is small, with 0.01 mg m^{-3} between the maximum and minimum, similar to the variation in roof measurements in Klaić et al. (2012) (~ 0.015 mg m^{-3}). However, the measurements were taken in spring and autumn so atmospheric conditions would be very different. Also, the measurements were above road level so the advection of more distant sources was dominant over the emissions from local traffic (Klaić et al. 2012). Low variation in the present study may be explained by the measurement station being far from the road.

There is more fluctuation in the Saturday data (Fig. 8c), presumably as traffic flows are less regular than on working days. The major increase in concentrations is delayed until around 12:00 LST as expected for leisurely rather than work related traffic. Similarly, the morning traffic peak in Barcelona was delayed by 1 hour on weekends compared to weekdays in (Viana et al. 2011). Unexpectedly, the peak concentration on Sundays 21:00 - 22:00 LST (Fig. 8d) (0.0448 ± 0.0269 mg m^{-3}) is higher than for the other days, the reason for this is unclear, but it is notable that its standard deviation is the highest for Sundays. However, the gradual increase throughout the daytime, without significant peaks may represent a gradual build-up of pollutants over the day as traffic flows are low. As Sundays were only 10 out of the 70 day measurement period, more Sunday data is required to fully assess diurnal variation.

Mean weekly variation (Fig 8c) shows the lowest average concentrations for Saturdays (0.0279 ± 0.0187 mg m^{-3}) and Sundays (0.0296 ± 0.0191 mg m^{-3}), as expected with reduced traffic flows and the peak value for Mondays (0.0402 ± 0.0286 mg m^{-3}) and Tuesdays (0.0402 ± 0.0288 mg m^{-3}). 0.0123 mg m^{-3} (31 %) between the maximum and minimum value. Winter variations in [PM 1] were even lower (1.5 %) weekdays compared to weekends from roof measurements University in Milan (Vecchi et al. 2004), this was explained by long atmospheric residence times of [PM 1]. As well as geographical differences in sources and conditions, the difference in monitor height may explain differences.

3.3.2 Temporal variations in indoor [PM]

For the 75 days of measurements the average indoor diurnal variation is relatively low (Fig. 9a.), the mean range of values across a 24 hour period is small with 0.0102 mg m^{-3} (-16%) between the mean maximum at 11:00 LST (0.0627 ± 0.053611 mg m^{-3}) and minimum (0.0525 ± 0.0489 mg m^{-3}) at 05:00 LST. Similarly, This lack of substantial fluctuation is comparable with low diurnal variation in laboratories in (Viana et al. 2011) despite the absolute values being much lower (0.0065 – 0.008 mg m^{-3}). Lower values in (Viana et al. 2011) may be explained by the laboratory being vacant and only indirectly affected by human activity re-suspending of PM or by the room building being more sealed from outdoor air than in the present study.

The weekday minimum (Fig 9.b) was 0.0476 ± 0.0386 mg m^{-3} at 04:00 LST and peaks occur throughout the day to a maximum of 0.0617 ± 0.0526 mg m^{-3} at 21:00 LST. The early morning increase may be attributed to several factors; the earliest increases may be associated with thermal uplift as the central is increased after 06:00. Whilst infiltration of morning traffic particles, enhanced infiltration and re-suspension from drafts due to doors opening and movement along in the hallways may account for the other morning peaks. Similarly, small size fractions infiltrated a residential building residential building in California, whilst opening doors lead to peaks in [PM 1] concentrations in a residential home in (Vette et al. 2001). This increase in indoor [PM 1] due to increased air exchange rate is only expected if there is not significant indoor source (Chen et al. 2011), as in this study. A weekday morning [PM 1] peak was also found in offices in a vacant laboratory in Barcelona (Viana et al. 2011), attributed to morning activity in the closest corridor. The other peaks fit well with the movement of people in the building, 14:00 for lunch (0.0570 ± 0.0483 mg m^{-3}), 18:00 (0.0559 ± 0.0440 mg m^{-3}) for staff leaving the building and 21:00 (0.0617 ± 0.0526 mg m^{-3}) for the last person leaving the building. The following reduction in concentrations throughout the night to the 04:00 LST minimum is comparable to the 5% reduction in [PM 1] after the work shift was over in (Viana et al. 2011), although here the reduction was larger (22 %).

The general trend for Saturdays is similar to the outdoor Saturday data which may represent infiltration. There is a marked decrease in [PM 1] for Saturday early mornings to a minimum of 0.0489 ± mg m^{-3} at 06:00 LST, presumably as the PM from Friday evenings settles in the absence of new sources. The increase to 0.0527 ± 0.0436 mg m^{-3} at 08:00 LST is likely due to entry of the duty staff member and peak around 11:00-12:00 may be due to staff coming to work or the duty staff member taking a break, other peaks are not accounted for but the 6pm peak may be due to people leaving the building later than the duty staff member. As expected, with an absence of indoor activities and external sources there is a substantial decrease in [PM 1] to 0.0251 ± 0.0323mg m^{-3} at 23:00 LST, the lowest mean value for the whole week, on Saturday evenings.

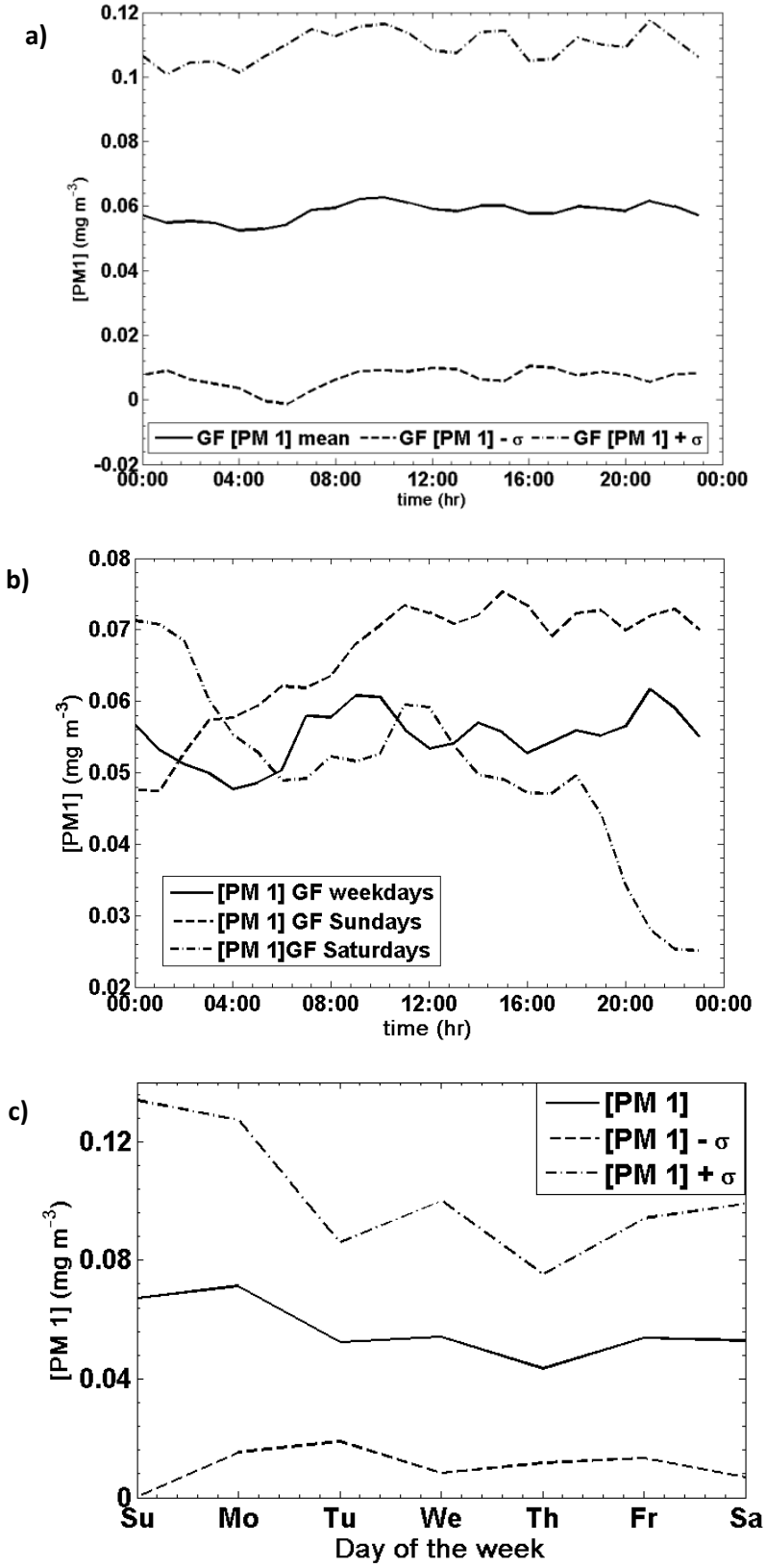

Fig. 9: Temporal variation in indoor PM 1 mass concentrations ([PM 1] (mg m^{-3})) 18.11.2012 – 30.01.2013. a) diurnal variation for the whole period, mean [PM 1] (solid line), mean + standard deviation (dashed line), mean – standard deviation (dash – dot line). b)Diurnal variation for weekdays (solid line), Saturdays (dash-dot line), Sundays and holidays (dashed line). c) Weekly variation in PM 1 mass concentrations ([PM 1] (mg m^{-3})) 18.11.2012 – 30.01.2013, Mean (solid line), mean + standard deviation (dashed line), mean – standard deviation (dash – dot line).

However, Sunday and holidays' high concentrations are difficult to account for with the highest maximum mean value for the week for 15:00 (0.0753 ± 0.0695 mg m^{-3}). But, the rise of [PM 1] indoors may be partially explained by the lack of ventilation on Sundays and holidays. As the windows are closed and usually no one enters the building. Without ventilation to remove particles, gradual heating of the inside throughout the day may cause thermal eddies to uplift particles. Furthermore, although concentrations were lower than in the present study, average [PM 1] was higher during weekend nights (0.00156 mg m^{-3}) in (Braniš et al. 2005) than for any other period during the week. The standard deviations are so large that it is clear that there are multiple controls on [PM 1].

Furthermore, analysis of Christmas holidays with Sunday data may cause some artefact as although the building should have been unoccupied during this period, traffic levels in the city during this period would reflect the normal work day pattern; so infiltration of pollutants may have increased the [PM 1] above that of a regular Sunday. Analysis of Sundays and holidays separately is recommended to check this. Moreover, the outdoor values for Sundays alone have higher than expected evening values so some higher than expected concentrations on Sundays may have affected this period. Aside from Sundays, values are lowest during the late night and early morning as in (Kearney et al. 2011). The [PM 1] fluctuation in this study and is attributed to dust re-suspension by workers and students passing through the building. Similarly, in (Viana et al. 2011) the main indoor [PM 1] source was worker movement within the building, this was confirmed by the high mineral content of [PM 1], characteristic of re-suspension, when workers were present in the building.

However, whilst larger size fractions were heavily influenced by human activity in classrooms in other studies (Vette et al. 2001; Braniš et al. 2005; Fromme et al. 2007; Goyal et al. 2009), [PM 1] was not substantially affected by human activity in the same investigations. Whilst the fluctuations in [PM 1] are generally not large in the present study, they coincide with the changes in other size fractions, as the majority of the larger fractions are made up of the smaller sizes in this study (section 3.5). Little difference was seen in the diurnal variation of the other size fractions (appendix 1). Fluctuations in all fractions coincide with the same time periods for [PM 1] but they are of slightly higher magnitude, as expected as they are more produced by mechanical means. It seems in the present study that [PM 1] is affected by human activity but not strongly. But to draw conclusions separate analysis of night and day time values is recommended to further investigate the impact of human activity and lack of ventilation on [PM 1]. Also, a time period could be investigated documenting human activity in the building alongside the measurements.

Mean weekly first floor [PM 1] is shown in Fig 9.c. The highest mean concentrations are apparent on Mondays, possibly explained due to re-suspension of dust after it settling during the weekend, infiltration of traffic pollution and the increase in building temperature from 17 to 21°C after the weekend causing thermal uplift of particles. The difference between the maximum and minimum is 39%. The highest daily mean is for Mondays (0.0713 ± 0.0777 mg m^{-3}), probably due to more traffic pollution in ambient air and re-suspension of PM in the building after a weekend of low activity. Whilst the lowest is for Thursdays (0.0435 ± 0.0496 mg m^{-3}), this is unexpected. Conversely, winter values were lowest at weekends in Braniš et al. (2009), and even lower during summer. As described for diurnal variation, high Sunday values are not fully understood. Again, for weekly variation the

standard deviations are very high, this suggests that temporal variation is not the only control on their variability.

3.4 Relationship between ambient and indoor [PM 1]

The relationship between inside and outside is shown in Fig.10, Fig.10a shows hourly mean [PM 1] from continuous inside and ambient measurements. The indoor and ambient concentrations appear to rise and fall simultaneously, whilst inside concentrations are generally higher. A closer look at the time series (Fig.10b) shows generally smoother variation in ground floor concentrations than outdoor. However, the peak in indoor data in the afternoon 21.11.2012 is not accounted for. The similarity of the curves suggests that the main control on indoor [PM 1] is the ambient concentration with an overprint of indoor sources, such as re-suspension or lack of removal of accumulated infiltrated PM due to low ventilation in winter as discussed in 3.3.2. Moreover, smoother variation in indoor [PM 1] may be explained by perturbation by human activity causing fluctuations on a smaller timescale, this resolution is lost in one hour averages. Similarly, ambient concentrations fluctuated more than indoor in Vette et al. (2001). The higher indoor [PM 1] are best related to the ambient concentration by a quadratic equation, but further analysis is required to provide an explanation of this relationship which has a strong correlation ($R^2 = 0.796$), (Fig. 10.c).

The average inside - outside ratio (I/O), calculated from calculating the I/O ratio of each hour and then finding the mean for the entire measurement period, was 1.64 (table 1). Yet I/O is highly variable with a minimum of 0, maximum 9.46 and standard deviation of 0.905. It is surprising that the I/O is greater than 1 as no [PM 1] sources were identified. I/O was less than 1 in a number of comparable studies of [PM 1] without significant indoor sources (e.g. Vette et al. 2001; Braniš et al. 2005; Sangiorgi et al. 2013). However, movement through offices was limited in Sangiorgi et al. (2013) and the measurements covered all seasons. Whilst Vette et al. (2001) covered winter and spring and most indoor activity was restricted to mornings. So seasonal differences and less of re-suspension may account for the lower I/O in both than in the present study. Furthermore, a strong diurnal pattern was observed, with stronger correlation between I/O at night (Vette et al. 2001). Whilst it was beyond the scope of this study to investigate the temporal variations in I/O, this would help determine the influence of indoor activities and ambient conditions which vary temporally and seasonally.

Conversely, I/O greater than 1 without significant indoor sources was also reported in classrooms in Poland (Polednik 2013), classrooms and homes in N. India (Goyal et al. 2009; Massey et al. 2012) and in supermarkets in Beijing (Brunekreef et al. 2005). However, it is difficult to make comparisons because coal combustion sources in (Polednik 2013) and generally higher pollution levels and substantially different climatic conditions in India and China. As with absolute concentrations, further inter study differences including measurement conditions, PM sources, building type and indoor activity make direct comparison of I/O difficult.

Possible causes for high I/O include building design and weather conditions. As this building was built in the 1980s and maintains its original windows, it is expected to be much less sealed than modern

buildings. So as well as entry of pollutants through open doors and windows, cracks in the building envelope allow infiltration of outdoor air (Chen et al. 2011), the temperature gradient in winter enhances the infiltration of cold air. Thus, in the absence of indoor and ventilation systems, pollutants may accumulate inside. Furthermore, some artefact in I/O may result from different measurement techniques for the indoor and outdoor data and also the location of the outdoor monitor not being directly outside the measurement site for indoor [PM 1] so concentrations may be lower than at the measurement location. Also, as discussed in section 3.2.2 changes in wind speed and direction may indirectly influence indoor concentrations by forcing more pollutants through the building envelope. For example, (Ní Riain et al. 2003) found the I/O in a University building in Central London was highest when winds were directed from the most polluted region to building facade, but this was for a naturally ventilated building and larger size fractions. For the present study further analysis of I/O ratios in relationship to wind direction and other climatic variables is recommended to further investigate controls on I/O. I/O investigations could be further improved by simultaneous measurement, using the same measurement methods at the same resolution, in and outside of the same building to avoid spatial and instrumental discrepancies in source location.

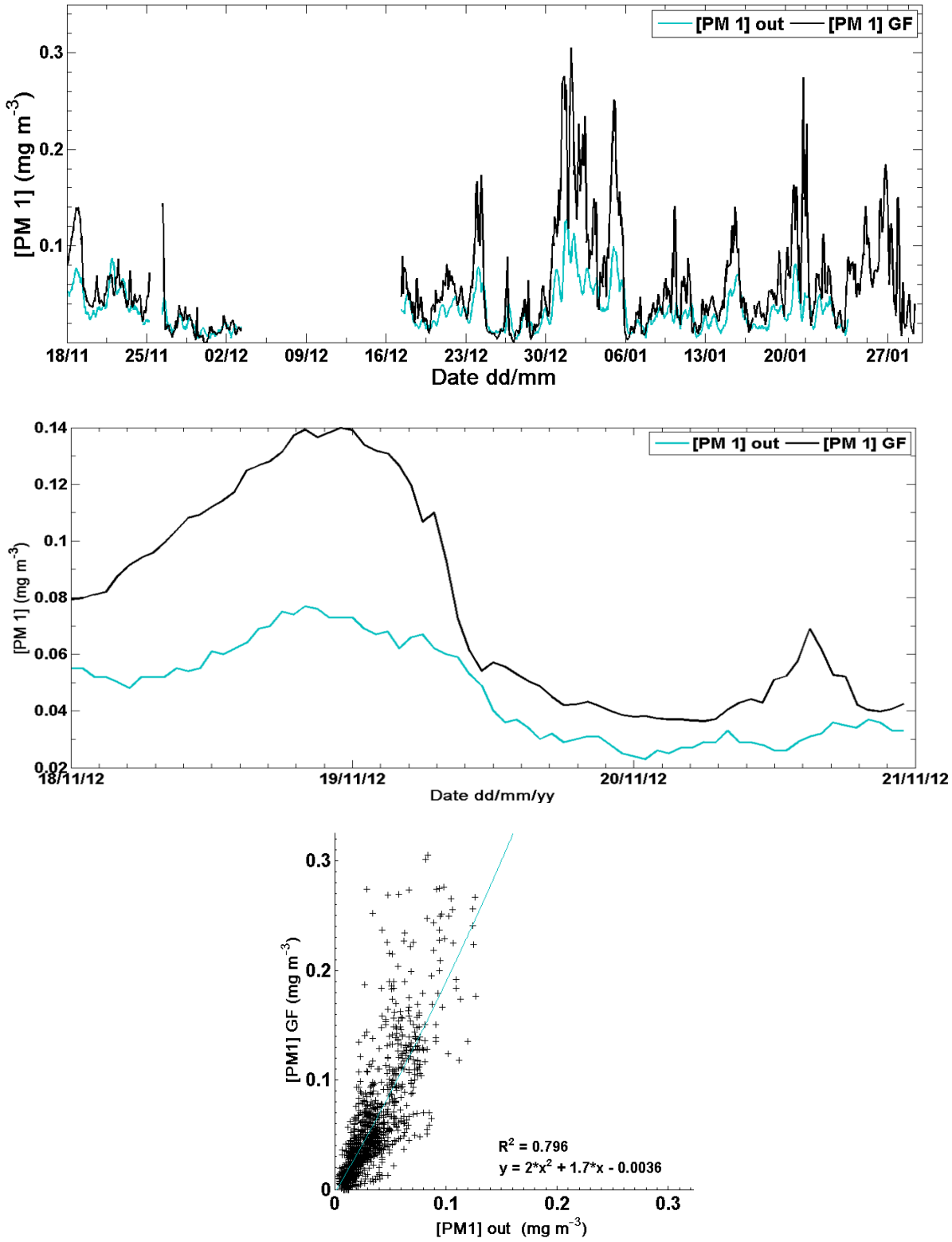

Fig. 10 Relationships between indoor ground floor and ambient PM mass concentrations ([PM 1] GF and [PM] out respectively, (mg m^{-3})). a) Time series of [PM 1]GF (black line) and [PM] out (blue line) from continuous, minute data 18.11.12 – 30.01.2012. b) time series of [PM 1]GF (black line) and [PM] out (blue line) from continuous, minute data 18.11.12 – 21.01.2012. c) scatter plot of hourly mean [PM 1]GF and [PM] out, with a quadratic trend line (blue line).

3.5. Individual size fraction contributions to indoor TSP.

Table 3: Ratios (Pearson's correlation coefficients) between different size fractions on the ground (GF) and first floor (FF). Ratios were calculated as (larger fraction)/(smaller fraction) to be comparable with the literature. P= 0.1 for all R^2 values.

	[PM 1] GF/	[PM 1] FF/	[PM 2.5]/	[PM 4]/	[PM 10]/	[TSP]/
/[PM 1] GF		1.165 (0.991)				
/[PM 1] FF	0.894 (0.991)		1.002 (1.000)	1.007 (1.000)	1.024 (0.999)	1.081 (0.991)
/[PM 2.5]				1.004 (1.000)		
/[PM 4]					1.017 (1.000)	
/[PM 10]						1.049 (0.996)

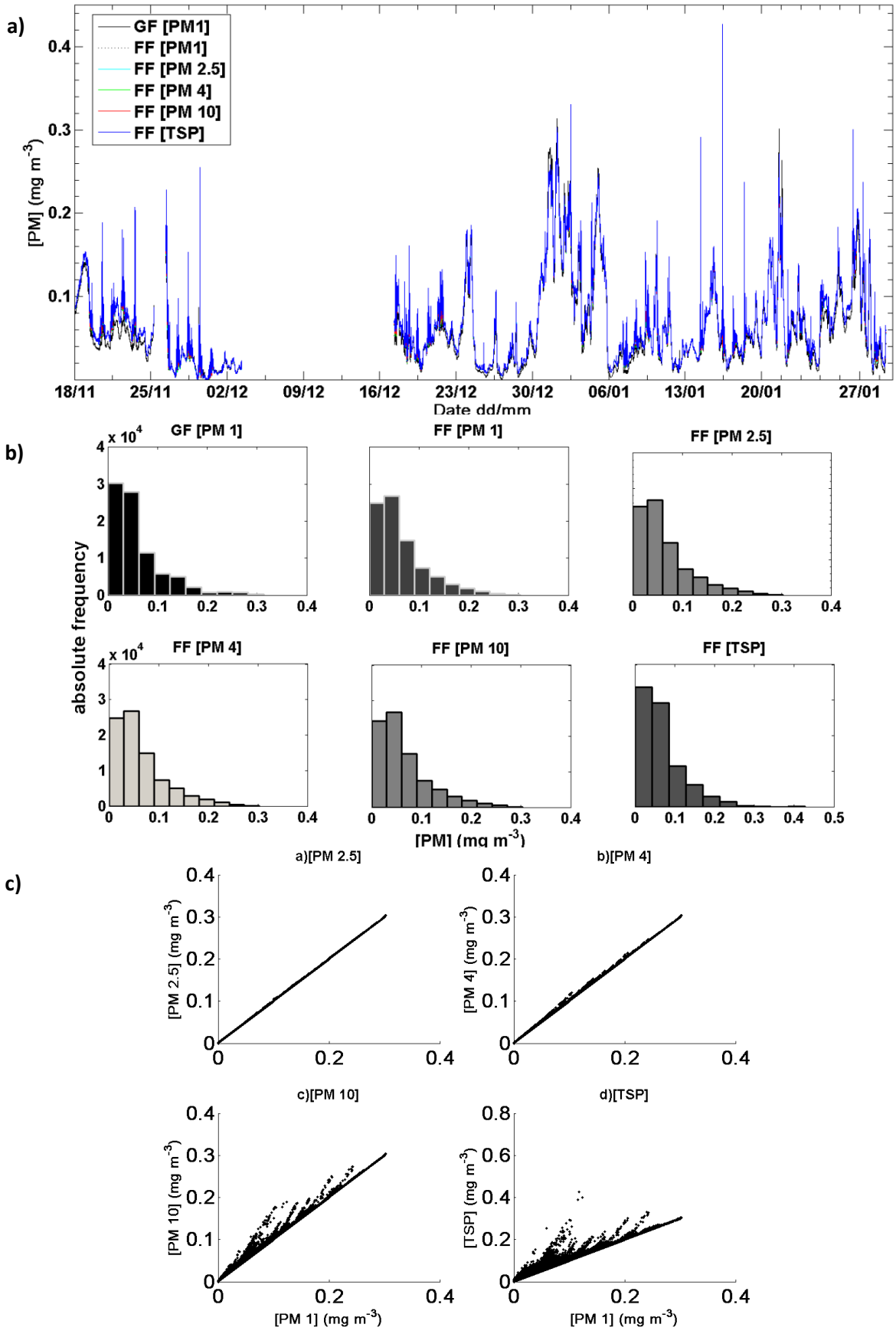

Fig. 11: PM size fraction relationships. a)times series of all [PM]. b) absolute frequency of all [PM]. c) size dependence of larger fractions on [PM 1].

In the time series for all indoor size fractions (Fig. 11a) the continuous variation in [PM] for size fractions are virtually indistinguishable. This is emphasised by Fig.11b showing that for all size fractions, the most frequent concentrations are below 0.1 mg m^{-3}. Whilst the very lowest concentrations (< 0.033 mg m^{-3}) are the most common in ground floor (GF) [PM 1], on the first floor (FF) those between 0.033 and 0.066 mg m^{-3} are more prominent. Also, in table 1 the mean values for the first floor size fractions only deviate by 0.0001-0.0005 mg m^{-3} (0.0633 – 0.641 mg m^{-3} for [PM 1] and [PM 10] respectively).

Fig 11.c shows the dependence of larger size fractions and TSP on [PM 1], there is not a noticeable difference between [PM 1] and [PM 2.5], this is expected as they both result from combustion in ambient conditions and are not created indoors. Whilst the larger fractions generated by different processes deviate away from [PM 1]. Yet, none of the size fractions deviate far from [PM 1] or each other as ratios and R^2 values for the between the size fractions are all close to 1 (table 3), higher values than found in the literature. As larger fractions are mainly associated with indoor sources (e.g. Jones et al. 2000; Braniš et al. 2009), there are no significant indoor sources in the present study, so it is unsurprising that their concentrations don't deviate far from the smaller fractions.

The variation in size fractions is much lower than for lecture rooms in Prague in winter (0.0137 - 0.0423 mg m^{-3} for [PM 1] and [PM 10] respectively) (Braniš et al. 2005) and classrooms in winter in Munich (0.0239 – 0.0719 mg m^{-3} [PM 2.5] and [PM 10] respectively) (Fromme et al. 2007). Mean ratios and correlation coefficients between size fractions in the present study are all close to 1 (table 3). Whilst ratios ranged 0.32 - 0.87 and R^2 0.762-0.991 for [PM 1]/[PM 10] on work day times and [PM 2.5]/[PM 10] for weekend nights respectively in (Braniš et al. 2005) and lower values were found in (Liu et al. 2004). However, much more human activity is expected the higher capacity university (Braniš et al. 2005), school (Fromme et al. 2007) and various public buildings (Liu et al. 2004) than in the building in the present study, as larger size fractions are more perturbed by human activity than in smaller ones, this may partly account for differences. This is supported by closer correlation between all size fractions when the building was empty in (Braniš et al. 2005), future work in this area could separate size fraction analysis into high and low activity periods to assess this relationship further.

3.6 First floor and Ground floor relationships.

In time series (Fig. 12.a.) calculated from continuous minute data, ground floor and first floor [PM 1] follow each other closely, the highest [PM 1] fluctuates between ground and first floor without a clear pattern, although first floor [PM 1] appears higher more frequently and the first floor ground floor ratio In table 3 is 1.165. But the relationship is not completely clear as the mean [PM 1] for ground floor is lower (0.0579 mg m^{-3}) than for the first floor (0.0633 mg m^{-3}) the maximum value is higher for ground floor and the median is lower for the first floor (table 1).

Slight predominance in first floor [PM 1] could be explained by higher activity on the first floor, as workers and students pass in and out of offices and lecture rooms. But further investigation of the differences in activity levels and diurnal variations in the ratio between first and ground floor would

need to be investigated to assess this further. Also, as measurements were taken during the heating season, thermal uplift of particles could result from temperature gradients between the first and ground floor. Times when the ground floor [PM 1] are higher are explained by being closer to the entrance of the building so worker entry allows infiltration of ambient air, more activity downstairs or gravitational settling of [PM 1] from the first floor. Again a further investigation of activity in the building in relationship to concentrations, diurnal variations and the relationship between air movement and concentrations are needed to assess this relationship further.

The relationship between the two is best described by a quadratic as the relationship tails off at higher concentrations showing a strong correlation between the two (R^2 = 0.990) (Fig. 12.b.). This was calculated from hourly means and the first floor predominance is not apparent. This suggests that perturbations are important only important in short temporal variation. Furthermore, box plots from minute data show a simpler positive linear relation (appendix 2). This close relationship between different spaces within the same building is similar to the correlation in [PM 1] between classrooms in the same school in Munich (R^2 = 0.93) (Fromme et al. 2007). Intra-room relationships were stronger in winter in (Fromme et al. 2007) so investigations in summer for the same conditions as the present study would help further evaluate spatial variation in [PM 1].

4. Conclusions and Recommendations

75 days of minute mean [PM] and meteorological variables and 70 days of ambient [PM 1] were analysed. Absolute concentrations of ambient [PM 1] (0.0339 ± 0.233 mg m^{-3}) were comparable with ambient measurements in Belgium (Berghmans et al. 2009) and Prague (Braniš et al. 2005). [PM 1] indoors (0.0579 ± 0.0515 mg m^{-3}) was similar to classrooms in Lublin, Poland (Polednik 2013). When compared to meteorological variables, ambient [PM 1] relationships generally agreed with the work of Klaić et al. (2012) with wind speed, T, RH and ρ showing the strongest relation with [PM 1]. Yet, the potential for strong seasonal variation in these relationships was identified. So an inter-seasonal study is suggested under the same measurement conditions. For indoor [PM 1] the strongest relationships were found with ambient T, RH and V_h, again further seasonal investigation would be beneficial.

Ambient [PM 1] temporal variations followed expected patterns in for traffic variations, with lower magnitude fluctuation than expected, explained by the position of the monitor. A maximum 30% change in [PM 1] was observed across the week and 50% across the diurnal cycle. Yet, unexpectedly there were high concentrations for Sunday afternoons. Similarly, indoor [PM 1] had the highest concentrations for Sundays and holidays, most likely explained by lack of ventilation whilst the building was empty. For the rest of the week diurnal variations were similar to outdoor and relatively low fluctuations, up 16 % for diurnal and 39% for weekly were detected. Indoor concentrations were likely related to a combination of infiltrated ambient air and meteorology, indoor activity perturbation, heating and ventilation.

Whilst the indoor-outdoor relationship showed concurrent rises and falls in concentration, the I/O ratio was high (1.64) and was highly variable. Compared to other studies without substantial indoor sources, I/O greater than 1 was found in classrooms in Poland (Polednik 2013), classrooms and homes in N. India (Goyal et al. 2009; Massey et al. 2012) and in supermarkets in Beijing (Brunekreef et al. 2005). But due to the differences in climate and outdoor sources, only tentative comparisons were made. Ratios lower than one were common in the literature (e.g. Vette et al. 2001; Braniš et al. 2005; Sangiorgi et al. 2013). But activity levels and seasonal differences may account for differences. With such high variability in I/O it is recommended that temporal variations in I/O are investigated to better understand their relationship between indoor activity, outdoor sources, meteorological variables. Furthermore, I/O investigations could be improved by simultaneous measurement in and outside of the same building to avoid spatial discrepancies in source location.

The majority of [PM] indoors was of the smallest size fractions. Ratios and R^2 values between larger sizes and PM 1 were all close to 1. Whilst ratios were lower in the literature, most likely explained by differences in activity levels and sources. Similarly, there was little difference in the ground and first floor [PM 1] with a mean ratio 0.894, comparable with inter room relationships in winter in (Fromme et al. 2007).

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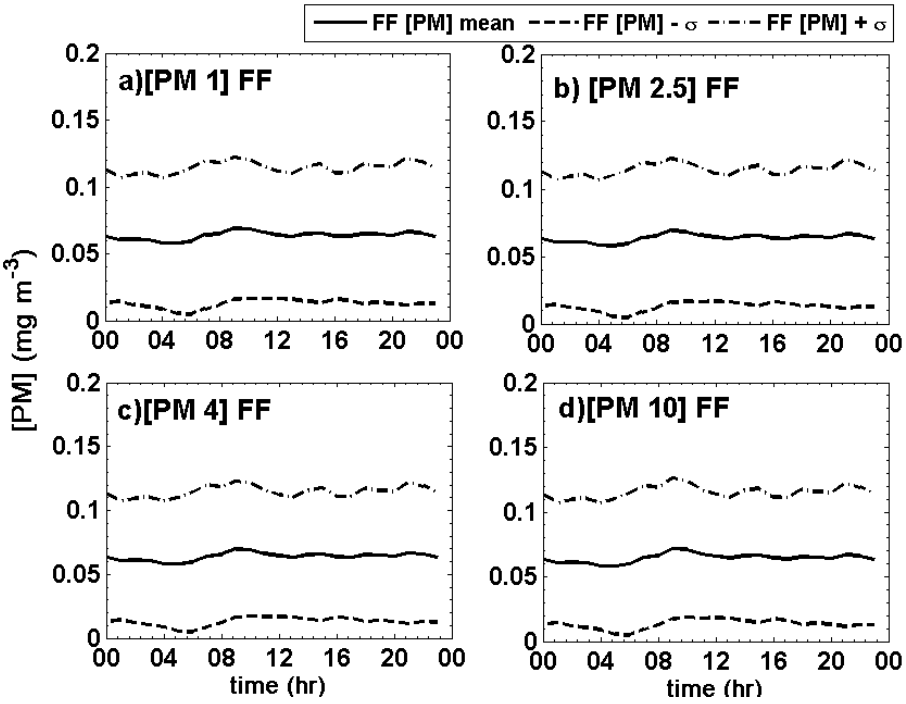
Appendices

Appendix 1



Appendix 2